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PROCESSING CERAMICS

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16. Abstract A method of hot hydrostatic pressing of ceramics is described. A detailed description of the invention is given. The invention is explained through an actual example, and a figure illustrates the temperature and pressure during HIP treatment. ORIGINAL FROM OF POOR QUALITY					
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54 Hot hydrostatic pressing of ceramics

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Specifications

1. Title of Invention

Hot hydrostatic pressing of ceramics.

2. Scope of Patent Claim

(1) A method of hot hydrostatic pressing of ceramic powder mold immersed in a glass bath through compression sintering using pressure-transmitting gas at a high temperature in which the entire surface of the aforementioned mold is covered with powder consisting of material which does not melt during hot hydrostatic pressing and which is sintered at a temperature lower than the sintering temperature of the aforementioned mold material with BN as the inner layer, this method characterized by sintering the aforementioned coating powder material in the course of the aforementioned hot hydrostatic pressing treatment to form an air-tight layer followed by heating to the sintering temperature of the aforementioned mold.

(2) The method of hot hydrostatic pressing of ceramics of Claim 1 in which the ceramic powder mold is a mold consisting primarily of silicon nitride, silicon carbide or boron carbide.

(3) The method of hot hydrostatic pressing of ceramics of Claims 1 and 2 in which the coating material is Al_2O_3 , ZrO_2 or BN.

(4) The method of hot hydrostatic pressing of ceramics of Claims 1, 2 and 3 in which the glass is pyrex glass.

3. Detailed Description of the Invention

This invention concerns hot hydrostatic pressing treatment using pressure-transmitting gas, especially a method of improving the sealing properties of the aforementioned hot hydrostatic pressing treatment through the using of glass with a low softening point in which pressure-transmitting gas is dissolved as sealant.

The method of hot hydrostatic pressing (below abbreviated HIP) using glass as the sealant has the advantages of forming different shaped molds and using copper or [illegible], which are ordinarily used as capsule material, even at high temperatures at which they could not be used. The glass capsule method (consult Patent Publication No. 46-2731) and the method of embedding mold in glass powder (consult Patent Disclosure No. 55-89405) are well known. At present, these are used extensively as high temperature structural material in molding processing of ceramic powder whose development has been progressing.

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However, heat resistant ceramic powder has a high sintering temperature. Since the HIP temperature is higher than the previous temperature during metal powder molding, the same state develops whether using the aforementioned glass sealing method or the glass bath method since the HIP temperature reaches 1700°C in treating ceramics such as silicon nitride (Si_3N_4) or silicon

carbide (SiC) using glass with a low softening point. In fact, the ceramic mold to be treated is immersed in a glass bath and it is covered with molten glass. Thus, in this case, part of the mold floats out of the glass bath since the specific gravity of the ceramic mold is 1.8 to 2.1 g/cm² until high density is reached, which is lower than the specific gravity of 2.2 to 2.3 of glass, and sealing is incomplete. This is due to the reaction between slight amounts of oxygen contained in the gas and the ceramic which is induced through the direct contact made between the pressure-transmitting gas and the mold when Al or N₂ gas is used as the pressure medium during HIP treatment.

In addition, when using pyrex glass, for example, as glass with a low softening point, the pressure-transmitting gas dissolves at specific temperatures and pressures, thereby resulting in the collection of this gas in the pores of the mold which results in internal pressure and the inability to achieve high density due to HIP treatment.

However, in spite of these defects, glass with a low softening point has the advantages of being easy to handle and being inexpensive. Thus, improvement of the aforementioned defects is desired because these advantages are extremely beneficial in industry.

The objective of this invention is to provide a method of improved sealing which can effectively utilize glass sealant, which is an extremely useful sealant, by overcoming the aforementioned defects of glass.

Specifically, the features of this invention which achieve the aforementioned objectives are realized through a method of HIP of ceramic powder mold immersed in a glass bath by compression sintering at a high temperature using air as the pressure medium in which the entire surface of the aforementioned mold is covered with powder consisting of material which is sintered at a temperature lower than the sintering temperature of the mold material with BN powder as the inner layer. This method is characterized by sintering the aforementioned coating material in the course of HIP treatment to form an air-tight layer followed by heating to the sintering temperature of the aforementioned mold.

The concrete modes of implementation of the aforementioned are explained below.

First, the HIP treatment method which is the basic method of treatment in this invention is a method of compression sintering in which ceramic powder mold is sealed in a glass capsule or is embedded in glass powder followed by compression sintering at a high temperature using an inert gas such as Ar or N₂ gas as the pressure medium while the ceramic is immersed in the glass bath.

The ceramic powder used here would be composed primarily of silicon nitride, silicon carbide or boron carbide (B₄C). It would be formed as a preliminary mold or as a preliminary sintered substance followed by sealing in a glass capsule or embedding in glass powder and HIP treatment.

The mold to be treated which has been subjected to preliminary molding or preliminary sintering may also contain sintering promoters such as Y_2O_3 powder, Al_2O_3 powder or MgO powder in addition to the ceramic powder itself.

Conversely, means of coating the aforementioned mold to be treated include sealing in glass capsules or embedding in glass powder, and the same state is exhibited upon heating in either case to a temperature above the softening point of glass. However, degasification is simple in the former case since only the degasified section is enclosed in the capsule in degasification sealing in order to avoid the effects of residual air upon sealing, while degasification of the entire heating furnace is necessary in the latter case, which constitutes some difference.

Possible examples of the glass used in this invention include silica glass, Vycor glass or pyrex glass, but pyrex glass is the most desirable since silica glass forms a reactive layer with the mold while Vycor glass is difficult to remove following treatment. HIP treatment is usually carried out by temperature- and pressure elevation in an atmosphere of Ar or N_2 etc. in the HIP furnace, but the temperature should first be elevated above the softening point of glass to form fluid glass while the atmospheric pressure is under 100 atmospheres in order to prevent the development of local strain by the glass capsule or the trapping of high pressure gas within the material to be treated through the formation of a dense glass layer on the outer surface

of the material through fusion of the glass powder, thereby making HIP treatment impossible. When glass powder is used, a dense glass layer forms on the outer surface of the treated material followed by temperature- and pressure elevation to the treatment and pressure levels set for HIP treatment. However, the aforementioned HIP process is not always appropriate since the coating material between the glass coating and the mold to be treated forms a sintered layer in this invention, as stated below.

Specifically, this invention forms a specific coating layer over the entire surface of the material to be treated which ensures complete sealing in the HIP treatment, as stated above.

Accordingly, the entire outer surface of the mold to be treated would be sintered at a temperature below the mold sintering temperature before sealing the aforementioned glass capsule or before embedding in glass powder, thereby completing a layer of powder material which is not melted even at the HIP treatment temperature.

Al_2O_3 , ZrO_2 or BN may be used as the coating material, and BN powder would be the most desirable among these, but the material is not restricted to BN powder because of the relation to the ceramic mold to be subjected to HIP treatment. However, BN powder has good mold releasing properties and a BN powder layer must be formed directly on the outer surface covering the mold to be treated. Accordingly, a BN powder layer is formed on the inside to facilitate mold release while coating material

which is characteristic of this invention is affixed to the outer surface.

While metal powder is not excluded from the coating material, the metals are restricted to Mo, Pt or Cr, but Cr presents difficulty in mold release following treatment.

Thus, these coating materials should be in a form such as a rubber flexible sealant as required in order to maintain the shape. The interior of the mold would be packed with coating material powder, and the required mold to be treated would be therein. A layer would be formed by hydrostatic treatment at normal temperature.

The thickness of one layer of the coating material should exceed 0.3 mm, preferably 0.5 mm. In any case, the coating material must form an air-tight sintered layer on the surface of the mold to be treated at a temperature of 1200°C preferably during HIP treatment, and the material must be suited for densification through HIP treatment.

The types of coating material, the temperature and pressure conditions in sintering until an air-tight layer is formed, the types of ceramic molds subjected to HIP treatment and the HIP temperature and pressure are given in the following table.

Coating material		
Al ₂ O ₃	1200°C	1000 kgf/cm ²
ZrO ₂	1350°C	1000 "
BN	1650°C	1800 "

Ceramic mold		
Si ₂ N ₄	1750°C	2000 kgf/cm ²
SiC	1850°C	"
B ₄ C	1850°C	"

The coating material would first be sintered at position A on the curve of pressure and temperature illustrated in the appended figure when selecting the aforementioned coating material for the material to be treated which has been selected from the aforementioned ceramic molds and conducting treatment thereof. This would be followed by sintering at position B of the mold to be treated which is covered with said coating material.

Accordingly, in treatment using pressure-transmitting gas in a glass bath based on the method of this invention, there are differences in the specific gravity, but the seal is complete even if some of the material were to float up from the glass bath during treatment. Thus, high densification can be expected due to suitable uniform compression in HIP treatment.

As indicated above, the method of this invention utilizes the advantage of utilization of glass with a low softening point as the sealant, thereby overcoming the defect of sealing properties. Thus, this invention has great industrial utility.

When using glass in which pressure-transmitting gas is dissolved, for example pyrex glass, as the sealant, compression sintering would be followed by revaporization of the pressure-transmitting gas in which glass is melted, thereby converting the glass into bubbling form. By cooling this, the appended glass

can be removed with slight mechanical external force, thereby getting two birds with one stone. /414

Below, this invention is explained through an actual example.

(Actual Example)

Si_3N_4 powder containing 2.5% Y_2O_3 with 70% α phase and mean particle diameter of approximately $1\mu\text{m}$ was subjected to uniform compression under pressure of $5,000 \text{ kgf/cm}^2$, thereby producing mold to be treated with relative density of 62%.

Bn was coated on the surface of this mold to a thickness of 0.3 mm followed by embedding in pyrex glass in a graphite crucible and insertion in an HIP device. A sintered state was achieved at a specific position (position A in appended figure) during HIP treatment since Al_2O_3 is sintered at 1200°C , $1,000 \text{ kgf/cm}^2$ as stated above. BN also was sintered at the aforementioned specified temperature and pressure through temperature elevation and pressure elevation. Finally, the temperature and pressure were elevated to 1750°C , 2000 kgf/cm^2 (position B in figure) which is the temperature and pressure for HIP treatment of Si_3N_4 mold.

The temperature was reduced to 500°C while the pressure was maintained at 2000 kgf/cm^2 following HIP treatment, and the pressure was then gradually released. A mold covered with glass was removed from the crucible, and this was then held for 30 minutes under heating to 1100°C in an electric furnace, followed by cooling.

The glass which covered the mold became bubbling glass due to vaporization of the pressure-transmitting gas which was dissolved in the glass, and the flakant could be removed very easily. The resulting mold was a high density sintered material with relative density of 98.5%, and the compression over the entire surface was uniform, resulting in a quality product.

4. Brief Description of Drawing

The figure illustrates the temperature and pressure during HIP treatment.

